

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Dieter KERNER *et al.*

Appl. No. unknown

Filed: February 28, 2002

For: SURFACE-MODIFIED, DOPED,
PYROGENICALLY PRODUCED
OXIDES

Art Unit: unknown

Examiner: n/a

Atty. Docket No. 39509-177800

Customer No.



26694

PATENT TRADEMARK OFFICE

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, DC 20231

Sir:

Please amend the following claims:

2. (Amended) Surface-modified, pyrogenically produced oxides doped by aerosol, characterized in that the oxides are selected from the group consisting of SiO_2 , Al_2O_3 , TiO_2 , B_2O_3 , ZrO_2 , In_2O_3 , ZnO , Fe_2O_3 , Nb_2O_5 , V_2O_5 , WO_3 , SnO_2 and GeO_2 .

3. (Amended) The surface-modified, pyrogenically produced oxides according to claim 1 or 2, wherein the surface-is modified with one or several compounds selected from the following groups:

a) Organosilanes of the type $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n+1})$ and $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n-1})_2$

$\text{R} = \text{alkyl}_1$

$n = 1 - 20;$

b) Organosilanes of the type $\text{R}'_x (\text{RO})_y \text{Si}(\text{C}_n\text{H}_{2n+1})$ and $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n+1})$

$\text{R} = \text{alkyl}_1$

$\text{R}' = \text{alkyl}_1$

$\text{R}' = \text{cycloalkyl}$

$\text{N} = 1 - 20,$

$x+y = 3,$

$x = 1, 2,$

$y = 1, 2;$

c) Halogen organosilanes of the type $\text{X}_3 \text{Si}(\text{C}_n\text{H}_{2n+1})$ and $\text{X}_3 \text{Si}(\text{C}_n\text{H}_{2n-1})$

$\text{X} = \text{Cl}, \text{Br},$

$n = 1 - 20;$

d) Halogen organosilanes of the type $\text{X}_2 (\text{R}') \text{Si}(\text{C}_n\text{H}_{2n+1})$ and

$\text{X}_2 (\text{R}') \text{Si}(\text{C}_n\text{H}_{2n-1}),$

$\text{X} = \text{Cl}, \text{Br}$

$\text{R}' = \text{alkyl}$

$R' = \text{cycloalkyl}$

$n = 1 - 20;$

e) Halogen organosilanes of the type $X (R')_2 \text{Si}(\text{C}_n\text{H}_{2n+1})$ and

$X (R')_2 \text{Si}(\text{C}_n\text{H}_{2n-1})$,

$X = \text{Cl, Br};$

$R' = \text{alkyl}$

$R' = \text{cycloalkyl}$

$n = 1 - 20;$

f) Organosilanes of the type $(\text{RO})_3\text{Si}(\text{CH}_2)_m\text{-R}'$

$R = \text{alkyl},$

$m = 0.1 - 20,$

$R' = \text{methyl-, aryl (e.g., -C}_6\text{H}_5, \text{ substituted phenyl groups)}$

$-\text{C}_4\text{F}_9, \text{OCF}_2\text{-CHF-CF}_3, -\text{C}_6\text{F}_{13}, -\text{O-CF}_2\text{-CHF}_2,$

$-\text{NH}_2, =\text{N}_3, -\text{SCN}, -\text{CH=CH}_2, -\text{NH-CH}_2\text{-CH}_2\text{-NH}_2,$

$-\text{N-(CH}_2\text{-CH}_2\text{-CH}_2\text{NH}_2)_2,$

$-\text{OOC(CH}_3)_c = \text{CH}_2,$

$-\text{OCH}_2\text{-CH(O)CH}_2,$

$-\text{NH-CO-N-CO- (CH}_2)_5,$

$-\text{NH-COO-CH}_3, -\text{NH-COO-CH}_2\text{-CH}_3, -\text{NH-(CH}_2)_3\text{Si(or)}_3,$

$-\text{S}_x\text{-(CH}_2)_3\text{Si(OR)}_3,$

-SH, and

-NR'R''R''', wherein R' = alkyl, aryl; R'' = H, alkyl, aryl; and R''' = H, alkyl, aryl, benzyl, or C₂H₄NR'''' with R'''' = H, alkyl and R'''' = H, alkyl;

g) Organosilanes of the type (R'')_x(RO)_ySi(CH₂)_m-R'

R'' = alkyl, cycloalkyl,

x+y = 2,

x = 1, 2,

y = 1, 2,

m = 0.1 to 20,

R' = methyl-, aryl, -C₆H₅, substituted phenyl groups

-C₄F₉, -OCF₂-CHF-CF₃, -C₆F₁₃, -O-CF₂-CHF₂,

-NH₂, -N₃, SCN, -CH=CH₂, -NH-CH₂-CH₂-NH₂,

-N-(CH₂-CH₂-NH₂)₂,

-OOC(CH₃)C=CH₂,

-OCH₂-CH(O)CH₂,

-NH-CO-N-CO-(CH₂)₅

-NH-COO-CH₃, -NH-COO-CH₂-CH₃, -NH-(CH₂)₃Si(OR)₃,

-S_x-(CH₂)₃Si(OR)₃

-SH, and

-NR'R''R''', wherein R' = alkyl, aryl; R'' = H,

alkyl, aryl; and R''' = H, alkyl, aryl, benzyl, or

C₂H₄NR'''' R'''''' with R'''' = H, alkyl and

R'''''' = H, alkyl) ;

h) Halogen organosilanes of the type X₃Si (CH₂)_m-R'

X = Cl, Br,

m = 0, 1 – 20,

R' = methyl-, aryl-, -C₆H₅, substituted phenyl groups

-C₄F₉, -OCF₂-CHF-CF₃, -C₆F₁₃, -O-CF₂-CHF₂,

-NH₂, -N₃, SCN, -CH=CH₂, -NH-CH₂-CH₂-NH₂,

-N-(CH₂-CH₂-NH₂)₂,

-OOC (CH₃)C = CH₂,

-OCH₂-CH(O) CH₂,

-NH-CO-N-CO-(CH₂)₅,

-NH-COO-CH₃, -NH-COO-CH₂-CH₃, -NH-(CH₂)₃Si(OR)₃,

-S_x-(CH₂)₃Si(OR)₃, and

-SH;

i) Halogen organosilanes of the type (R)₂Si(CH₂)_m-R'

X = Cl, Br,

R = alkyl such as methyl, - ethyl-, propyl-,

m = 0, 1 – 20,

R' = methyl-, aryl [(e.g.), -C₆H₅, substituted phenyl groups,

-C₄F₉, -OCF₂-CHF-CF₃, -C₆F₁₃, -O-CF₂-CHF₂,

-NH₂, -N₃, SCN, -CH=CH₂, -NH-CH₂-CH₂-NH₂,

-N-(CH₂-CH₂-NH₂)₂,

-OOC (CH₃)C = CH₂,

-OCH₂-CH(O) CH₂,

-NH-CO-N-CO-(CH₂)₅,

-NH-COO-CH₃, -NH-COO-CH₂-CH₃, -NH-

(CH₂)₃Si(OR)₃,

-S_x-(CH₂)₃Si(OR)₃ ,

-SH;

(j) Halogen organosilanes of the type (R)₂X Si(CH₂)_m-R'

X = Cl, Br,

R = alkyl,

m = 0, 1 – 20,

R' = methyl-, aryl-, -C₆H₅, substituted phenyl groups,

-C₄F₉, -OCF₂-CHF-CF₃, -C₆F₁₃, -O-CF₂-CHF₂,

-NH₂, -N₃, SCN, -CH=CH₂, -NH-CH₂-CH₂-NH₂,

-N-(CH₂-CH₂-NH₂)₂,

-OOC (CH₃)C = CH₂,

-OCH₂-CH(O) CH₂,

-NH-CO-N-CO-(CH₂)₅,

-NH-COO-CH₃, -NH-COO-CH₂-CH₃, -NH-(CH₂)₃Si(OR)₃,

-S_x-(CH₂)₃Si(OR)₃

-SH;

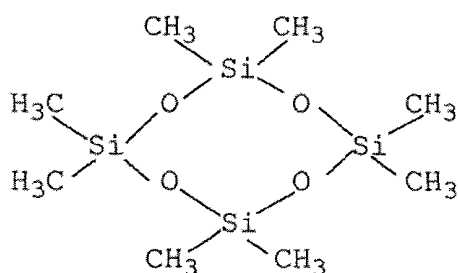
(k) Silazanes of the type $R'_2Si-N-SiR'_2$



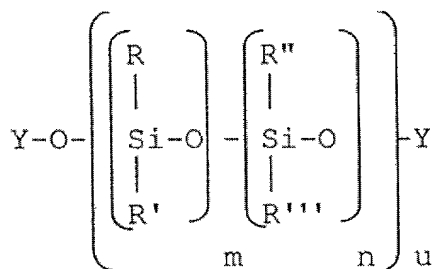
R = alkyl,

R' = alkyl, vinyl; or

(l) Cyclic polysiloxanes of the type D 3, D 4 or D 5:



m) Polysiloxanes or silicone oils of the type



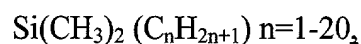
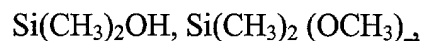
$$m = 0, 1, 2, 3, \dots \infty$$

$$n = 0, 1, 2, 3, \dots \infty$$

$$u = 0, 1, 2, 3, \dots \infty$$

$$Y=CH_3, H, C_nH_{2n+1} \quad n=1-20$$

$$Y=Si(CH_3)_3, Si(CH_3)_2H$$



R = alkyl, aryl, $(CH_2)_n-NH_2$, H,

R' = alkyl, aryl, $(CH_2)_n-NH_2$, H,

R'' = alkyl, aryl, $(CH_2)_n-NH_2$, H,

R''' = alkyl, aryl, $(CH_2)_n-NH_2$, H.,

4. (Amended) A method of producing the surface-modified oxides in accordance with claim 1 or 2, comprising placing pyrogenically produced oxides doped by aerosol in a suitable mixing container, spraying the oxides under intensive mixing with the surface-modification reagent or a mixture of several surface-modification reagents.

5.(Amended) In a reinforcing filler composition wherein the improvement comprises the surface-modified oxides according to claim 1 or 2 as reinforcing filler.

6. (New) The method of claim 4 wherein the spraying step includes spraying with water and/or acid prior to the spraying with the surface-modification reagent or a mixture of several surface-modification reagents.

7. (New) The method of claim 4 further comprising re-mixing at 15 to 30 minutes and tempering at a temperature of 100 to 400 °C for a period of 1 to 6 hours.

8. (New) The surface-modified, pyrogenically produced oxides according to claim 3 wherein the cyclic polysiloxanes is type D 4.

9. (New) The surface-modified, pyrogenically produced oxides according to claim 8 wherein the type D4 cyclic polysiloxanes is octamethylcyclotetrasiloxane.

ABSTRACT:

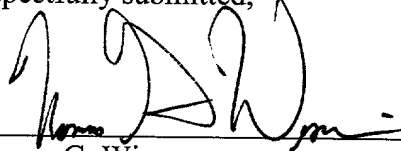
Surface-modified, doped, pyrogenically produced oxides surface-modified by spraying pyrogenically produced oxides doped by aerosol under intensive mixing with a surface-modification reagent or a mixture of several surface-modification reagents. These reagents include organosilanes, silazanes, cyclic polysiloxanes, polysiloxanes or silicone oils. The organosilanes include: $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n+1})$, $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n-1})$; $\text{R}'_x(\text{RO})_y\text{Si}(\text{C}_n\text{H}_{2n+1})$; $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n+1}) \text{X}_3 \text{Si}(\text{C}_n\text{H}_{2n+1})$, $\text{X}_3 \text{Si}(\text{C}_n\text{H}_{2n-1})$; $\text{X}_2(\text{R}') \text{Si}(\text{C}_n\text{H}_{2n+1})$, $\text{X}_2(\text{R}') \text{Si}(\text{C}_n\text{H}_{2n-1})$; $\text{X}(\text{R}')_2 \text{Si}(\text{C}_n\text{H}_{2n+1})$; $\text{X}(\text{R}')_2 \text{Si}(\text{C}_n\text{H}_{2n-1})$; $(\text{RO})_3\text{Si}(\text{CH}_2)_m\text{-R}'$; $(\text{R}'')_x(\text{RO})_y\text{Si}(\text{CH}_2)_m\text{-R}'$; $\text{X}_3\text{Si}(\text{CH}_2)_m\text{-R}'$; $(\text{R})\text{X}_2\text{Si}(\text{CH}_2)_m\text{-R}'$, and $(\text{R})_2\text{XSi}(\text{CH}_2)_m\text{-R}'$. The silazanes include $\text{R}'\text{R}_2\text{Si-N-Si(H)R}_2\text{R}'$. The surface-modification permits the pyrogenically-produced oxides to be more rapidly worked into organic systems, e.g. polyester resins, at higher concentrations.

REMARKS

It is respectfully submitted that the present claims are in condition for examination on the merits. However, if any questions remain, the Examiner or other PTO official is encouraged to call the undersigned at (202) 513-4614 expedite this application.

In the unlikely event that the transmittal letter is separated from this document and the Patent Office determines that an extension and/or other relief is required, applicant petitions for any required relief including extensions of time and authorizes the Assistant Commissioner to charge the cost of such petitions and/or other fees due in connection with the filing of this document to **Deposit Account No. 22-0261**. However, the Assistant Commissioner is not authorized to charge the cost of the issue fee to the Deposit Account.

Respectfully submitted,



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APPENDIX

Amended claims only (with markings to show changes)

1. Surface-modified, pyrogenically produced oxides doped by aerosol.

2. (Amended) Surface-modified, pyrogenically produced oxides doped by aerosol, characterized in that the oxides are [oxides] selected from the group consisting of SiO_2 , Al_2O_3 , TiO_2 , B_2O_3 , ZrO_2 , In_2O_3 , ZnO , Fe_2O_3 , Nb_2O_5 , V_2O_5 , WO_3 , $\text{SnO}_{2[\text{,}]}$ and GeO_2 .

3. (Amended) The surface [Surface]-modified, pyrogenically produced oxides [doped by aerosol in accordance with] according to claim 1 or 2, [characterized in that they are] wherein the surface-is modified with one or several compounds selected from the following groups:

a) Organosilanes of the type $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n+1})$ and $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n-1})_2$

$\text{R} = \text{alkyl}_1$

$n = 1 - 20_2$

b) Organosilanes of the type $\text{R}'_x(\text{RO})_y\text{Si}(\text{C}_n\text{H}_{2n+1})$ and $(\text{RO})_3\text{Si}(\text{C}_n\text{H}_{2n+1})$

$\text{R} = \text{alkyl}_1$

$\text{R}' = \text{alkyl}_1$

$\text{R}' = \text{cycloalkyl}$

$\text{N} = 1 - 20_2$

$$x+y=3_i$$

$$x=1, 2_i$$

$$y=1, 2_i$$

c) Halogen organosilanes of the type $X_3 \text{ Si}(\text{C}_n\text{H}_{2n+1})$ and $X_3 \text{ Si}(\text{C}_n\text{H}_{2n-1})$

$$X = \text{Cl}, \text{Br}_i$$

$$n = 1 - 20_i$$

d) Halogen organosilanes of the type $X_2 (\text{R}') \text{ Si}(\text{C}_n\text{H}_{2n+1})$ and

$$X_2 (\text{R}') \text{ Si}(\text{C}_n\text{H}_{2n-1})_{\Delta}$$

$$X = \text{Cl}, \text{Br}$$

$$\text{R}' = \text{alkyl}$$

$$\text{R}' = \text{cycloalkyl}$$

$$n = 1 - 20_i$$

e) Halogen organosilanes of the type $X (\text{R}')_2 \text{ Si}(\text{C}_n\text{H}_{2n+1})$ and

$$X (\text{R}')_2 \text{ Si}(\text{C}_n\text{H}_{2n-1})_{\Delta}$$

$$X = \text{Cl}, \text{Br}_i$$

$$\text{R}' = \text{alkyl}$$

$$\text{R}' = \text{cycloalkyl}$$

$$n = 1 - 20_i$$

f) Organosilanes of the type $(RO)_3Si(CH_2)_m-R'$

$R = \text{alkyl}$

$m = 0.1 - 20$

$R' = \text{methyl-, aryl (e.g., } -C_6H_5, \text{ substituted phenyl groups)}$

$-C_4F_9, OCF_2-CHF-CF_3, -C_6F_{13}, -O-CF_2-CHF_2$

$-NH_2, =N_3, -SCN, -CH=CH_2, -NH-CH_2-CH_2-NH_2,$

$-N-(CH_2-CH_2-CH_2NH_2)_2$

$-OOC(CH_3)_c = CH_2$

$-OCH_2-CH(O)CH_2$

$-NH-CO-N-CO-(CH_2)_5$

$-NH-COO-CH_3, -NH-COO-CH_2-CH_3, -NH-(CH_2)_3Si(or)_3$

$-S_x-(CH_2)_3Si(OR)_3$

$-SH$, and

$-NR'R''R'''$,wherein $[(R' = \text{alkyl, aryl; } R'' = H, \text{ alkyl, aryl; and } R''' = H, \text{ alkyl, aryl,$

benzyl, or $C_2H_4NR''''R'''''$ with $R'''' = [A]H$, alkyl and

$R'''' = H, \text{ alkyl;}$

g) Organosilanes of the type $(R'')_x(RO)_ySi(CH_2)_m-R'$

$R'' = \text{alkyl, cycloalkyl,}$

$x+y = 2$

$x = 1, 2$

$y = 1, 2$

$m = 0.1$ to 20 ,

$R' =$ methyl-, aryl [(e.g.), $-C_6H_5$, substituted phenyl groups[]]

$-C_4F_9$, $-OCF_2-CHF-CF_3$, $-C_6F_{13}$, $-O-CF_2-CHF_2$,

$-NH_2$, $-N_3$, SCN , $-CH=CH_2$, $-NH-CH_2-CH_2-NH_2$,

$-N-(CH_2-CH_2-NH_2)_2$,

$-OOC(CH_3)C=CH_2$,

$-OCH_2-CH(O)CH_2$,

$-NH-CO-N-CO-(CH_2)_5$

$-NH-COO-CH_3$, $-NH-COO-CH_2-CH_3$, $-NH-(CH_2)_3Si(OR)_3$,

$-S_x-(CH_2)_3Si(OR)_3$

$-SH$, and

$-NR'R''R'''$, wherein [($R' =$ alkyl, aryl; $R'' = H$,

alkyl, aryl; and $R''' = H$, alkyl, aryl, benzyl, or

$C_2H_4NR''''R'''''$ with $R'''' = [A]H$, alkyl and

$R'''' = H$, alkyl);

h) Halogen organosilanes of the type $X_3Si(CH_2)_m-R'$

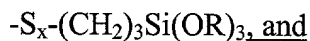
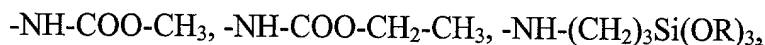
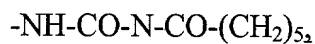
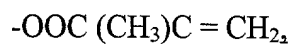
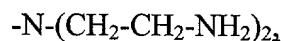
$X = Cl, Br$,

$m = 0, 1 - 20$,

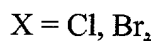
$R' =$ methyl-, aryl [(e.g.), $-C_6H_5$, substituted phenyl groups[]]

$-C_4F_9$, $-OCF_2-CHF-CF_3$, $-C_6F_{13}$, $-O-CF_2-CHF_2$,

$-NH_2$, $-N_3$, SCN , $-CH=CH_2$, $-NH-CH_2-CH_2-NH_2$,



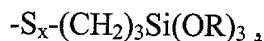
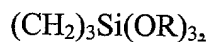
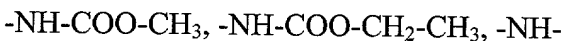
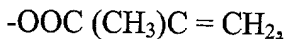
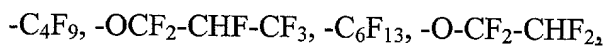
i) Halogen organosilanes of the type $(\text{R})\text{X}_2\text{Si}(\text{CH}_2)_m\text{R}'$



R = alkyl such as methyl, - ethyl-, propyl-,

m = 0, 1 – 20,

R' = methyl-, aryl [(e.g.), -C₆H₅, substituted phenyl groups[]],



-SH₂

(j) Halogen organosilanes of the type (R)₂X Si(CH₂)_m-R'

X = Cl, Br₂

R = alkyl₁

m = 0, 1 - 20₂

R' = methyl-, aryl [(e.g.), -C₆H₅, substituted phenyl groups[]],

-C₄F₉, -OCF₂-CHF-CF₃, -C₆F₁₃, -O-CF₂-CHF₂₂

-NH₂, -N₃, SCN, -CH=CH₂, -NH-CH₂-CH₂-NH₂,

-N-(CH₂-CH₂-NH₂)₂₂

-OOC (CH₃)C = CH₂₂

-OCH₂-CH(O) CH₂₂

-NH-CO-N-CO-(CH₂)₅₂

-NH-COO-CH₃, -NH-COO-CH₂-CH₃, -NH-(CH₂)₃Si(OR)₃₂

-S_x-(CH₂)₃Si(OR)₃

-SH₂

(k) Silazanes of the type R'R₂Si-N-SiR₂R'

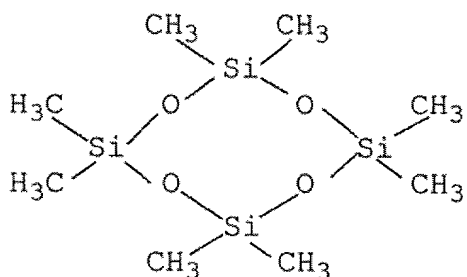
|
H

R = alkyl₁

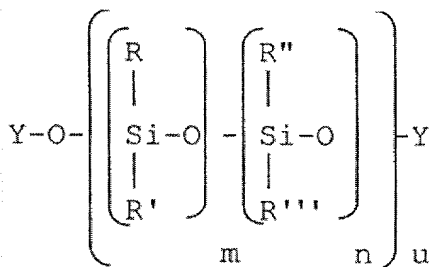
R' = alkyl, vinyl; or

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(l) Cyclic polysiloxanes of the type D 3, D 4[, or D 5[, e.g. octamethylcyclotetrasiloxane = D4]:



m) Polysiloxanes or silicone oils of the type



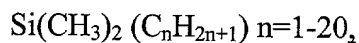
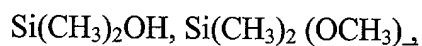
$$m = 0, 1, 2, 3, \dots \infty$$

$$n = 0, 1, 2, 3, \dots \infty$$

$$u = 0, 1, 2, 3, \dots \infty$$

$$Y=CH_3, H, C_nH_{2n+1} \quad n=1-20$$

$$Y=Si(CH_3)_3, Si(CH_3)_2H$$



R = alkyl, aryl, $(CH_2)_n-NH_2$, H ,

R' = alkyl, aryl, (CH₂)_n-NH₂, H₂

R'' = alkyl, aryl, (CH₂)_n-NH₂, H₂

R''' = alkyl, aryl, (CH₂)_n-NH₂, H₂

4. (Amended) A method of producing the surface-modified oxides in accordance with claim 1 or 2, [characterized in that] comprising placing pyrogenically produced oxides doped by aerosol [are placed] in a suitable mixing container, spraying the oxides [are sprayed] under intensive mixing[, optionally with water and/or acid at first and subsequently] with [a] the surface-modification reagent or a mixture of several surface-modification reagents,[optionally re-mixed 15 to 30 minutes and tempered at a temperature of 100 to 400 °C for a period of 1 to 6 hours].

5.(Amended) [The use of] In a reinforcing filler composition wherein the improvement comprises the surface-modified oxides according to claim 1 or 2 as reinforcing filler.

6. (New) The method of claim 4 wherein the spraying step includes spraying with water and/or acid prior to the spraying with the surface-modification reagent or a mixture of several surface-modification reagents.

7. (New) The method of claim 4 further comprising re-mixing at 15 to 30 minutes and tempering at a temperature of 100 to 400 °C for a period of 1 to 6 hours.

8. (New) The surface-modified, pyrogenically produced oxides according to claim 3 wherein the cyclic polysiloxanes is type D 4.

9. (New) The surface-modified, pyrogenically produced oxides according to claim 8
wherein the type D4 cyclic polysiloxanes is octamethylcyclotetrasiloxane.

[illegible]